2.0   MATERIALS AND METHODS

2.1   Overview

The test house was procured for use in April 1999 and configured for use in the study. Teflon® sampling lines were routed from the attached garage laboratory to the house for collection of gaseous pollutants (CO, NO, and NO_2). A second set of polypropylene sampling lines was installed for collection of the SF_6 that was measured to calculate air exchange rates. The sampling lines were approximately 15 m long. Signal lines were routed from the laboratory to the house for collection of data from the room temperature sensors, RH probes, thermocouples, and the power transducer. Lines were also routed for collection of outdoor temperature and RH. Pollutant monitors, the data acquisition system (DAS), and support hardware were set up in the garage laboratory.

Following set up and calibration of the instrumentation, testing began in the house. A used gas range was procured for the study, and a gas line was installed for the range. An electric range was also procured because the one in the house when rented did not have a self-cleaning feature. Preliminary measurements were performed to validate performance of the monitoring instrumentation. Air exchange rates were measured over a few days to validate performance of the method. The pollutant monitors were operated to measure background CO, NO, and NO_2 concentrations indoors and outdoors. Tests were then performed to measure baseline CO, NO, and NO_2 concentrations during operation of the gas range top burner and the oven without food cooking. Following the initial measurements, adjustments were made to the gas range burners, and additional measurements were performed.

The pre-test was performed in May 1999. It involved measurements of all parameters during two types of cooking activities – frying of loose ground beef in an un-covered pan on the gas range top burner and baking a pork roast in the gas oven. Both types of cooking were performed in duplicate in order to evaluate the variability between tests. Duplicate samples were collected during the tests to determine method precision. Results of the tests were used to refine the study design.

The main study was performed in February 2000. Thirty-two cooking tests were performed. Additional tests were performed to measure baseline concentrations of pollutants generated by the gas range top burner and the oven. The following sub-sections describe the main study.

2.2   Description of the Test House and Appliances

A test house was rented in Rohnert Park, California for use in the project. Rohnert Park, located approximately 60 miles north of San Francisco and 20 miles from the Pacific Ocean has a relatively moderate climate. During February and March 2000, while the tests in the main study were being performed, the average high temperatures were 61.6 °F (16.4 °C) and 64.4 °F (18 °C), respectively. Average lows were 40.2 and 41.4 °F (4.6 and 5.3 °C), respectively for February and March 2000. The average heating degree-days for February is 15 (base 65 °F) for the years 1931 to 2000.

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The house, built in 1983, was a single story ranch style home on a crawl space. The floor plan of the house is depicted in Figure 2-1, which also depicts the sampling locations in the kitchen, living room, and master bedroom. The front elevation of the house is depicted in Figure 2-2. There were a number of criteria defined for selection of the house including the following:

- Single family residence of typical California construction
- Built after 1988
- Two to three bedrooms
- Single story
- Floor area of 1300 to 2000 ft^2
- Double glazed windows and adequate insulation to have air exchange rates representative of California residences
- Central heating and air conditioning system
- Located in moderate climate (to minimize the need for operating the heating or air conditioning system during testing
- Located in a residential area away from major industrial sources of PM and SVOC contaminants
- No fugitive dust sources nearby (gravel driveways or roadways)
- Located at least one mile away from establishments that emit cooking fumes,
- Range top exhaust that vents to outdoors
- Attached garage (to facilitate set up of the laboratory)

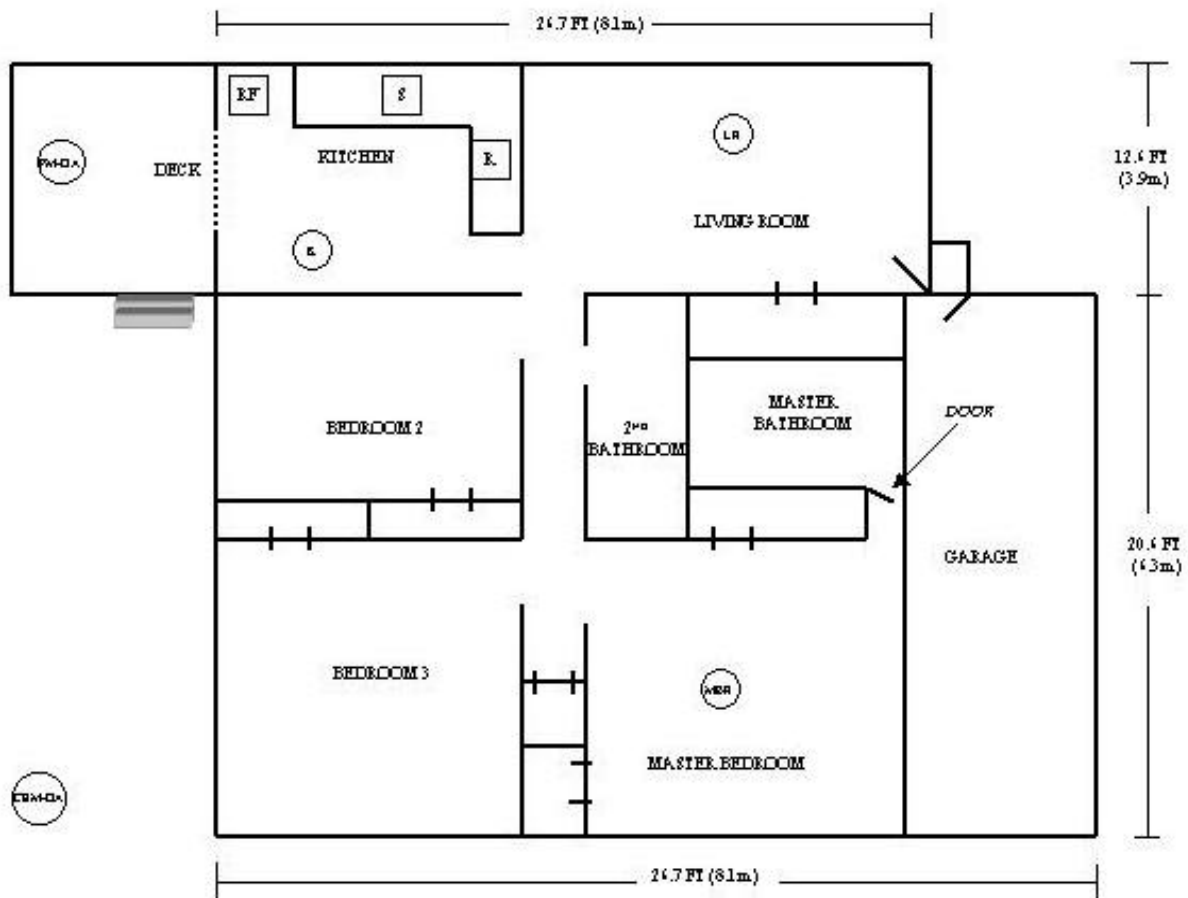Figure 2-1. Floor Plan of the Test House (Not to Scale)

Figure 2-2. Front Elevation of the Test House

Many of the criteria were met. However, some criteria were not met due to difficulty finding a rental home at the time that the study was performed. At the time of the study, there was an extremely limited rental home market in the area selected for the study. It was necessary to locate the test house in an area with a relatively moderate climate because the intention was not to operate a ducted heating and cooling system while tests were being performed. Operation of an air handler would likely result in substantial particle loss in the ductwork. There was difficulty finding homes that could meet the selection criteria.   There was also reluctance by landlords to rent a house for a one-year period for use as a test house. As a result of these complications, the search for an appropriate house that would meet the criteria lasted over five months.

The house that was rented was a single story home typical of California housing stock, but it was somewhat older and smaller than desired. The best available information is that it was built in 1983. The house had a floor area of 824 ft^2 (76.56 m^2) and a volume of 187 m^3. The size of the house was smaller than the original selection criterion. The rental company advised us that the house had a floor area of approximately 1100 ft^2, which turned out to be incorrect. However, as the data indicate, the small size and the layout of the house apparently resulted in good air mixing and relatively uniform air contaminant concentrations in the different rooms, most likely improving the accuracy of the source emission rate calculations. The house had three bedrooms, a living room, and kitchen separated by walls from the other rooms in the house (Figure 2-1). It had central heating, but no air conditioning. The furnace was fitted with a standard disposable high performance filter that was replaced at the start of the tests. The air exchange rate measurements showed that the house was relatively well insulated and sealed. Air exchange rates measured in

February 2000 ranged from 0.18 to 1.08 hr^{-1} and were typically 0.22 to 0.30 hr^{-1} (Table 3-25, Section 3). The range hood exhaust was vented to the outdoors. There were no major sources of particles from cooking activities located near the house. There was some wood burning in the residential neighborhood during the period of the main study.

The house was minimally furnished. There was a refrigerator in the kitchen, which was operating during the study. The furnishings in the adjacent living room consisted of an upholstered couch, upholstered loveseat, plastic chairs, and a coffee table. There were beds, but no dressers in two of the three bedrooms. There were plastic vertical blinds on all windows, but no curtains. The entire house, except for the bathrooms and kitchen, was carpeted. Therefore, although minimally furnished, there was a fairly substantial amount of surface area that could serve as reversible or non-reversible "sinks" for deposition of gaseous and particulate air contaminants. Plastic runners were placed over carpets in the main walkways of the living room and hallway in order to minimize particle resuspension by the technicians.

The house was equipped with an electric range when rented, but the oven did not include a self-cleaning feature. Therefore, a used electric range was obtained from a local appliance dealer. Gas piping was installed to the kitchen, and a gas range was purchased for the study. A microwave was also rented. All appliances were used and were at least five years old. The appliances were typical of those that would be found in a rental home of this size and age. All cooking appliances were cleaned prior to the start of testing. The only available information on the appliances was the following:

- Magic Chef Gas Range with self-cleaning oven, broiler in oven, and 4 equal-size, stovetop burners, Model # 34428RA (Figure 2-3). The right front burner was used for all range top burner tests. All of the burners were used in the full meal-cooking test.
- Whirlpool Electric Range with self-cleaning oven, 2 large burners, 2 small burners, Model No. RF385PXPW, Serial No. F62602406. The right front burner was used for all tests.
- Quasar Microwave Oven, Model No. MQ5540WW, Serial No. NM33409260, manufacture date of Dec. 1993, Output Frequency = 2450 MHz, Input = 110 volts, 13 amps.

During the main study, the main gas valve on the gas range failed after the first four tests. The valve was replaced. No other adjustments or changes were made to the gas range. The change of the valve did not impact the performance of the gas range top burners or oven.

A limited number of cooking utensils were purchased from local retail outlets in Rohnert Park, CA. They consisted of the following:

- Frying pan – Ecko Endura. 12-inch diameter, non-stick stainless steel skillet
- Baking dish – Pyrex, 9-in X 12-in
- Baking pan – Disposable aluminum baking pan, 8-in X 11-in
- Deep fat frying – Cast iron pan with aluminum frying basket (for French fries)
- Stir-frying–wok, carbon steel, 12 in. diameter
- Vegetable steaming and water boiling – 4 quart stainless steel steamers

Figure 2-3. Test House Gas Range and Cooking Utensils

2.3   Description of the Tests and Cooking Protocols

The main study consisted of the 32 cooking tests listed in Table 2-1. The tests involved cooking with a gas range, electric range, and microwave oven. Tests were performed to measure emissions for a variety of types of cooking, including frying of foods on the range top burners, broiling foods in the oven, and cooking meat in the oven. The tests addressed emissions from cooking of a variety of foods.

During the development of the study design, an attempt was made to obtain data on the predominant food types and cooking methods used in California. Population-based surveys of cooking method frequencies in California or the U.S. were not publicly available. Data on the dietary intake of the U.S. population is available from The U.S. Department of Agriculture (USDA) Continuing Survey of Food Intake of Individuals (CSFII) studies (USDA, 2000). But data on the dietary intake of Californians would be difficult to obtain from the CSFII database cost-effectively and within the time frame of this study. The CSFII studies show that beef, pork, and chicken are the most commonly consumed meats, and that vegetable and seed oils are a significant portion of the dietary fat intake in the U.S. Other information that was available suggested that, after baking or roasting meat, frying was the most common cooking method for meat (Mitchell, 1998). There was a high frequency of frying in fat and oils. Results reported by Rogge (1997) and Gerstler et al. (1998) showed that frying of food and broiling meat and fish resulted in high emissions.

Table 2-1. List of Cooking Tests

Test No.	Type	Range	Conditions	Notes/Comments[a]
1	Oven Cleaning	Gas	Standard	Oven problem; data not reported
1R	Oven Cleaning	Gas	Standard	Repeated test 1
2	Stovetop Stir Fry	Gas	Standard	Two batches cooked in one event
3A	Bacon	Gas	Standard	Two event cooking test
3B	Bacon			Second event
4	Tortillas	Gas	Standard	
5	French Fries	Gas	Standard	Two batches cooked in one event
6A	Broil Fish	Gas	Standard	Two event cooking test
6B	Broil Fish			Second event
7	Bake Lasagna	Gas	Standard	
8	Oven Cleaning	Electric	Standard	Tests 1–7 and 8–14 = same cooking protocols
9	Stovetop Stir Fry	Electric	Standard	Two batches cooked in one event
10A	Bacon	Electric	Standard	Two event cooking test
10B	Bacon			Second event
11	Tortillas	Electric	Standard	
12	French Fries	Electric	Standard	Two batches cooked in one event
13A	Broil Fish	Electric	Standard	Two event cooking test
13B	Broil Fish			Second event
14	Bake Lasagna	Electric	Standard	
15	Bacon	Microwave	Standard	Two event cooking test
15A	Bacon			Second event
16	Bake Lasagna	Microwave	Standard	
17A	Stovetop Stir Fry	Gas	Worst Case	Oil hotter; food cooked longer
17B	Stovetop Stir Fry			Second event in the test
18A	Bacon	Gas	Worst Case	Extra crisp bacon
18B	Bacon			Second event in the test
19A	Broil Fish	Gas	Worst Case	Burned
19B	Broil Fish			Second event in the test
20	Full Meal	Gas	Standard	Large meal with multiple foods; oven and range top burner used
21A	Stovetop Stir Fry	Gas	Test 2 Replicate	Not true replicate – changed to two event test
21B	Stovetop Stir Fry			Second event in the test
22	French Fries	Gas	Test 5 Replicate	
23	Bake Lasagna	Gas	Test 7 Replicate	
24	Fry Beef	Gas	Cast Iron Pan	Data logger failed
24RA	Fry Beef	Gas	Cast Iron Pan	Repeated Test 24 to evaluate pan type
24RB				Second event in the test
25	Pork Roast	Gas	Aluminum Pan	Compare pan types

Table 2-1. List of Cooking Tests Cont.

Test No.	Type	Range	Conditions	Notes/Comments[a]
26	Fry Beef	Gas	Range Hood	Operated on high
27	Pork Roast	Gas	Range Hood	Operated on high
28	Fry Beef	Gas	Range Side Shields	Exhaust on high with side shields
29	Pork Roast	Gas	Range Side Shields	Exhaust on high with side shields
30A	Stovetop Stir Fry	Gas	Vegetable Oil	Compare oils
30B				Second event in the test
31A	Fry Beef	Gas	Pan Lid	Impact of pan lid
31B				Second event in the test
32A	Popcorn	Microwave	Standard	Two event test
32B				Second event in the test
33	Burner Baseline	Gas		Range top burner with pot of water only
34	Oven Baseline	Gas		No food in oven
35	Instrument Comparison	Gas		No food in oven
36	Instrument Comparison	Gas		No food in oven
37	Instrument Comparison	Gas		No food in oven
38	Oven Dirtying	Gas		Bake off food and oil used to dirty oven
38R	Oven Dirtying	Gas		Bake off food and oil used to dirty oven
39	Oven Dirtying	Electric		Bake off food and oil used to dirty oven

[a] See text for definition of "test," "event," and "batches."

Based on the limited available information, discussions with the ARB, and teleconferences with members of the TRG, the matrix of tests presented in Table 2-1 was developed. The types of cooking included frying of foods on the range top because of the continued widespread use of frying for food preparation and the expected high emissions. Stir-frying of chicken and vegetables and frying of tortillas were expected to be common in the ethnically diverse cuisine and population of California. One type of cooking involved frying of loose ground beef in a pan on the range top burner because this food item can be used in a wide variety of food dishes. Broiling of fish in the oven was selected because it is a relatively simple food preparation method and likely to be common in California. A fish with high oil content, salmon, was selected for the broiling tests. The primary oven baking method involved cooking a large (2.5 lb.) pre-packaged lasagna, representing another easy type of cooking. Selection of the pre-packaged lasagna also allowed comparison of the emissions from gas and electric ovens with those from the microwave. Similarly, bacon was selected for the tests because it is a popular food item, and a comparison could be made between the microwave cooking method and standard frying on the range top. In a limited number of tests, a pork roast was cooked in the gas oven, allowing comparison of emissions from the meat with emissions from the lasagna. French fries were prepared by frying in oil because of the popularity of the food item and anticipation of high emissions from the heated oil. Two different cooking oils, vegetable and peanut oil, were used in the tests for comparison of emissions, particularly the PAH emissions. The oils were analyzed for PAH content prior to the main study.

The first 14 tests consisted of seven tests with the gas range for comparison to an identical set of seven tests with the electric range. Identical cooking protocols were used for the comparisons. Three tests were performed with the microwave. Two of the tests compared cooking of the same foods (bacon and lasagna) that were prepared with the gas and electric ranges. The other test was with a popular microwave food – popcorn. Three tests (Nos. 21, 22, and 23) were performed with the gas range as replicates of test numbers 2, 5, and 7, respectively.

The other tests were designed to evaluate factors that might impact emissions and indoor air concentrations during cooking. Tests 24 and 25 compared cooking with different pan materials. Test 24 was repeated as 24R because of a problem with the DAS during the first test. Tests 26 through 31 evaluated simple exposure reduction methods.

All tests involved preparation of a single food item, except test 20, which involved preparation of a full meal. All tests, except Test 20, were performed using either a range top burner or the oven, but not both at the same time. Each test, except Test 20, was performed for only a single type of cooking activity (e.g., broiling fish, stovetop stir-frying, baking a lasagna). In Test 20, preparation of the full meal involved cooking a number of different foods, using both the range top burners and the oven.

Also shown in the table are eight other tests that were performed. The gas range top burner was operated for one hour to obtain the baseline concentrations of combustion pollutants in Test 33. The oven was operated for two hours in Test 34 to obtain baseline concentrations. Tests 35, 36, and 37 were performed to evaluate sampling instrumentation in the kitchen. The last three tests listed (38, 38R, and 39) were not actually tests. For oven cleaning tests, the oven was first "dirtied" by applying a mixture of tomato puree and cherry pie filling to the bottom of the oven and cooking oil on the walls of the oven. After dirtying the oven, the oven was operated for a one-hour period at 350 °F to bake the material onto the surfaces in preparation for the test with the self-cleaning oven.

Tests 1 and 8 were performed to measure emissions during the self-cleaning cycles of the gas and electric ovens, respectively. Although oven cleaning is not performed frequently, the self-cleaning feature of both gas and electric ovens involves baking the surfaces at high temperatures (~450 °F). Therefore, this feature may result in high emissions from very dirty ovens. The Underwriters Laboratory (Underwriters Laboratory, 1993) has a method for dirtying an oven for measuring emissions of CO that involves application of 5-ounce of a beef gravy and vegetable oil shortening mixture. In January 2000, the Consumers Union reported a method for dirtying ovens during their performance tests of self-cleaning ovens (Consumers Reports, 2000). It involved a mixture of cherry pie filling, egg yolks, cheese, lard, tapioca, and tomato puree, but the quantities and test protocol could not be obtained in time for the tests. A modified protocol using five oz. of tomato puree, oil, and cherry pie filling was used in the first test, but the amount of material was too high, resulting in excessively high emissions. The amount was reduced to 3 oz. for tests 1R and 8. The protocol for dirtying the oven used a mixture of 28 g of tomato puree, 28 g of cherry pie filling, and 28 g of vegetable oil. The tomato puree and pie filling were wiped on the floor of the oven to represent spills. The oil was wiped on the walls of the oven with a brush. After application, the

oven was operated at 350 °F for one hour to bake the material onto the surface. The self-cleaning test was performed on the following day.

To the extent possible, cooking was performed following published recipes and routine cooking practices that would be performed in a residence. When available, as for the pre-packaged lasagna, the instructions on the package were followed. The cook, an ARCADIS chemist, was an experienced cook, but not a professional. He did much of the cooking in his own home and was considered to be representative of an "average" cook. By design, a professional cook was not used for the tests. During tests with the same food and cooking method, protocols were followed to insure that the food preparation methods, cooking methods, and cooking times were as consistent as possible to facilitate comparisons between tests.

As shown in Table 2-1, there were four types of cooking protocols implemented during the study:

1. Single event cooking tests with one batch of food - The simplest type of cooking protocol was a single event with a single batch of food cooked, as in Test Nos. 7 and 14 in which a single, large, pre-packaged frozen lasagna was baked in the oven. The cooking period was approximately two hours, and the post-exposure period was one hour, for a total exposure period of three hours.

2. Single event cooking tests during which two batches of food were prepared - In the second type of cooking protocol, there was a single event during which two batches of food were cooked sequentially. After all of the food was cooked, there was a one-hour post-cooking exposure period. This protocol was used in Test Nos. 5 and 12 during which a basket of French fries was cooked in oil, removed, and then a second batch was placed in the same oil to cook. It took approximately one hour to complete the cooking event, including the initial heating of the oil. After both batches of French fries were cooked, there was a one-hour post-cooking period, resulting in a total exposure period of approximately two hours.

3. Two event cooking tests - The third type of cooking protocol involved two cooking events separated by one hour. The protocol was used, for example, in Test Nos. 6 and 13. The test started by turning on the oven, then broiling a large salmon steak. At the end of the cooking period, the fish was removed and the oven was turned off. This cooking event, denoted as 6A in Table 2-1, was followed by a one-hour post-cooking period to simulate eating and clean-up. Then the oven was turned on again; a second salmon steak was broiled, and the oven was turned off. This cooking event was followed by another one-hour post-cooking period. The total duration of the test was 3.25 hours. The $PM_{2.5}$ and PM_{10} samples for gravimetric analysis were collected throughout the entire two-event cooking test. Continuous monitoring with the ELPI and other pollutant monitors provided concentration data over the entire test.

4. Exposure reduction tests consisted of replicate cooking experiments conducted with the addition of pan lid, range hood fan and side shields.

The need to perform these different protocols was demonstrated in the pre-test. During the pre-test, a single batch of loose ground beef was fried in a pan on the range-top burner. Due to the

small amount of food cooked and the short duration of the test, there was insufficient $PM_{2.5}$ and PM_{10} mass on the filters for accurate gravimetric analysis. To increase the amount of mass collected, these protocols were implemented.

The durations of the cooking events are presented in Table 2-2. The table lists the start time for background measurements, the cooking times, and the duration of the cooking time and the total exposure period. The total exposure period represents the total duration of the test. The test durations ranged from 1 hour and 30 minutes (Test 2) to 5 hours and 8 minutes for oven cleaning (Test 1R). The cooking period started when the range top burner or the oven was turned on. The cooking period ended when the burner or oven was turned off. Therefore, cooking periods included the time required to pre-heat the oven, pan, or cooking oil. The total exposure period was the total period in which the cook or occupants would be exposed to emissions from the cooking. The total exposure period included the cooking period plus a one-hour post-cooking period during which the meal would be eaten and cleanup would be performed. The assumption was that the cook and occupants would be in the residence for at least one hour after cooking. For the first two types of cooking protocols, the total exposure period included one hour of post-cooking exposure. In two event tests, the third type of protocol, there were two hour-long post-cooking exposure periods. A single $PM_{2.5}$ and PM_{10} mass measurement was made for each test regardless of the number of batches of food cooked or the number of cooking events within the test. During two event tests, data collected with the continuous monitors (CO, NO, NO_2, and PM with the ELPI) are reported as Test XA and XB in the results tables. The two event tests facilitated additional replication of cooking events, as recommended by the TRG, for measurements of CO, NO, NO_2, and PM with the ELPI.

Multiple cooking tests were performed on many days during the main study. Because of the amount of time necessary for zero and span checks of the continuous monitors, ELPI, and gas chromatograph used for SF_6 measurements, as well as set up of the instrumentation, performance of multiple tests on a day improved efficiency.

In order to obtain sufficient sample mass for PM, PAHs, and elements, a single set of outdoor air samples was collected on each day of testing. If only a single test was performed, for example with a total exposure period duration of 4 hours, the outdoor sample was collected during the same 4-hr period. If three tests were performed over a 9-hr period during the day, the outdoor air sample was integrated over the entire nine hours. This protocol was used because the mass of PM collected during short cooking tests would be inadequate for accurate gravimetric analysis.

On days with multiple tests, the house was flushed with outdoor air between tests to exhaust the air pollutants generated during the test and to re-establish the indoor background pollutant concentrations to reasonable levels prior to the start of the next test. The flush out was accomplished with the central air handler fan operating in conjunction with window fans and open doors. Because of this protocol, data was collected during most tests for only one hour after the source was turned off.

Cooking protocols were developed that involved reasonable and representative cooking scenarios. The detailed cooking protocols are included in Appendix C of the report. Table 2-3 presents a brief description of the cooking protocols. To the extent possible, cooking was

performed to be representative of typical residential cooking. All foods were purchased from local grocery stores in Rohnert Park. The foods to be used in multiple tests were purchased in large quantities and stored in the freezer to insure comparability between the tests. All pans, foods, and oils were weighed prior to, and following cooking, in order to determine the weight loss during the cooking event. Food weights are reported in Section 3.0. All cooking tests, except Test No. 20, involved cooking a single meal item. The stovetop stir-fry cooking protocol, for example, involved cooking both the vegetables and chicken for the dish, but involved use of only a single stovetop pan and burner. Most other cooking protocols involved a single food.

Table 2-2. Cooking Times and Test Durations

Test No.	Date	Test Type	Range Type	Cooking Event	Background Start Time	Cooking Start Time	Cooking End Time	Exposure End Time	Cooking Duration (Hr:Min)	Exposure Duration (Hr:Min)
1	2/3/00	Oven Cleaning	Gas	Total	12:44	12:54	16:54	18:24	4:00	5:30
1R	2/24/00	Oven Cleaning	Gas	Total	12:08	12:18	16:18	17:26	4:00	5:08
2	2/5/00	Stovetop Stir Fry	Gas	Total	13:59	14:09	14:30	15:39	0:21	1:30
3	2/5/00	Bacon	Gas	Total	9:08	10:08	11:57	13:06	0:42	2:58
3A	02/05/00	Bacon	Gas	1st event	9:08	10:08	10:30	11:36	0:22	1:28
3B	02/05/00	Bacon	Gas	2nd event	11:27	11:37	11:57	13:06	0:20	1:29
4	2/4/00	Tortillas	Gas	Total	10:31	10:41	11:26	12:38	0:45	1:57
5	2/4/00	French Fries	Gas	Total	13:31	13:41	14:49	16:13	1:08	2:32
6	2/6/00	Broil Fish	Gas	Total	10:54	11:04	12:55	14:05	0:44	3:01
6A	02/06/00	Broil Fish	Gas	1st event	10:54	11:04	11:28	12:34	0:24	1:30
6B	02/06/00	Broil Fish	Gas	2nd event	12:25	12:35	12:55	14:05	0:20	1:30
7	2/9/00	Bake Lasagna	Gas	Total	9:52	10:02	12:08	13:11	2:06	3:09
8	2/10/00	Oven Cleaning	Electric	Total	10:06	10:16	14:16	15:23	4:00	5:07
9	2/14/00	Stovetop Stir Fry	Electric	Total	14:02	14:12	14:33	15:44	0:21	1:32
10	2/11/00	Bacon	Electric	Total	15:06	15:23	17:14	18:25	0:51	3:02
10A	02/11/00	Bacon	Electric	1st event	15:06	15:23	15:46	16:45	0:23	1:22
10B	02/11/00	Bacon	Electric	2nd event	16:36	16:46	17:14	18:25	0:28	1:39
11	2/11/00	Tortillas	Electric	Total	9:52	10:02	10:45	11:54	0:43	1:52
12	2/11/00	French Fries	Electric	Total	12:47	12:57	13:22	14:35	0:25	1:38

Table 2-2. Cooking Times and Test Durations Cont.

Test No.	Date	Test Type	Range Type	Cooking Event	Background Start Time	Cooking Start Time	Cooking End Time	Exposure End Time	Cooking Duration (Hr:Min)	Exposure Duration (Hr:Min)
13	2/14/00	Broil Fish	Electric	Total	9:43	9:53	11:51	13:08	0:36	3:15
13A	02/14/00	Broil Fish	Electric	1st event	9:43	9:53	10:11	11:32	0:18	1:39
13B	02/14/00	Broil Fish	Electric	2nd event	11:23	11:33	11:51	13:08	0:18	1:35
14	2/13/00	Bake Lasagna	Electric	Total	15:26	15:36	17:37	18:58	2:01	3:22
15	2/8/00	Bacon	Microwave	Total	17:34	17:44	20:16	21:23	0:81	3:39
15A	02/08/00	Bacon	Microwave	1st event	17:34	17:44	18:28	19:38	0:44	1:54
15B	02/08/00	Bacon	Microwave	2nd event	19:29	19:39	20:16	21:23	0:37	1:44
16	2/8/00	Bake Lasagna	Microwave	Total	14:22	14:32	15:07	16:08	0:35	1:36
17	2/29/00	Stovetop Stir Fry	Gas	Total	14:50	15:00	16:56	18:38	0:51	3:38
17A	02/29/00	Stovetop Stir Fry	Gas	1st event	14:50	15:00	15:26	16:30	0:26	1:30
17B	02/29/00	Stovetop Stir Fry	Gas	2nd event	16:21	16:31	16:56	18:38	0:25	2:07
18	2/27/00	Bacon	Gas	Total	10:53	11:03	13:04	14:10	0:57	3:07
18A	02/27/00	Bacon	Gas	1st event	10:53	11:03	11:29	12:32	0:26	1:29
18B	02/27/00	Bacon	Gas	2nd event	12:23	12:33	13:04	14:10	0:31	1:37
19	2/27/00	Broil Fish	Gas	Total	18:45	18:55	20:49	21:57	0:50	3:02
19A	02/27/00	Broil Fish	Gas	1st event	18:45	18:55	19:20	20:23	0:25	1:28
19B	02/27/00	Broil Fish	Gas	2nd event	20:14	20:24	20:49	21:57	0:25	1:33
20	2/28/00	Full Meal	Gas	Total	8:38	8:48	11:08	15:53	2:20	7:05
21	2/27/00	Stovetop Stir Fry	Gas	Total	15:02	15:12	16:51	18:00	0:39	2:48

Table 2-2. Cooking Times and Test Durations Cont.

Test No.	Date	Test Type	Range Type	Cooking Event	Background Start Time	Cooking Start Time	Cooking End Time	Exposure End Time	Cooking Duration (Hr:Min)	Exposure Duration (Hr:Min)
21A	02/27/00	Stovetop Stir Fry	Gas	1st event	15:02	15:12	15:31	16:30	0:19	1:18
21B	02/27/00	Stovetop Stir Fry	Gas	2nd event	16:21	16:31	16:51	18:00	0:20	1:29
22	2/26/00	French Fries	Gas	Total	12:27	12:37	14:05	15:32	1:28	2:55
23	2/23/00	Bake Lasagna	Gas	Total	16:11	16:21	18:22	19:30	2:01	3:09
24	2/17/00	Fry Beef	Gas	Total	9:59	10:09	10:30	11:47	0:21	1:38
24R	2/25/00	Fry Beef	Gas	Total	10:20	10:30	12:13	13:25	0:42	2:55
24RA	2/25/00	Fry Beef	Gas	1st event	10:20	10:30	10:51	11:51	0:21	1:21
24RB	2/25/00	Fry Beef	Gas	2nd event	11:42	11:52	12:13	13:25	0:21	1:33
25	2/17/00	Pork Roast	Gas	Total	12:52	13:02	16:02	17:16	3:00	4:14
26	2/15/00	Fry Beef	Gas	Total	11:55	12:05	12:26	13:33	0:21	1:28
27	2/15/00	Pork Roast	Gas	Total	14:47	14:57	17:58	19:09	3:01	4:12
28	2/16/00	Fry Beef	Gas	Total	12:13	12:23	12:44	13:50	0:21	1:27
29	2/16/00	Pork Roast	Gas	Total	14:28	14:38	17:38	18:45	3:00	4:07
30	2/29/00	Stovetop Stir Fry	Gas	Total	10:32	10:42	12:17	13:29	0:35	2:47
30A	02/29/00	Stovetop Stir Fry	Gas	1st event	10:32	10:42	11:00	11:59	0:18	1:17
30B	02/29/00	Stovetop Stir Fry	Gas	2nd event	11:50	12:00	12:17	13:29	0:17	1:29
31	2/23/00	Fry Beef	Gas	Total	11:56	12:06	13:50	14:58	0:39	2:52
31A	02/23/00	Fry Beef	Gas	1st event	11:56	12:06	12:27	13:31	0:21	1:25
31B	02/23/00	Fry Beef	Gas	2nd event	13:22	13:32	13:50	14:58	0:18	1:26
32	2/15/00	Popcorn	Microwave	Standard	20:05	20:15	21:18	22:19	0:08	2:04

Table 2-2. Cooking Times and Test Durations Cont.

Test No.	Date	Test Type	Range Type	Cooking Event	Background Start Time	Cooking Start Time	Cooking End Time	Exposure End Time	Cooking Duration (Hr:Min)	Exposure Duration (Hr:Min)
32A	2/15/00	Popcorn	Microwave	1st batch	20:05	20:15	20:19	21:13	0:04	0:58
32B	2/15/00	Popcorn	Microwave	2nd batch	21:03	21:13	21:17	22:19	0:04	1:06
33	2/24/00	Range Baseline	Gas	Total	8:51	9:01	10:01	11:05	1:00	2:04
34	2/24/00	Oven Baseline	Gas	Total	18:15	18:25	20:25	21:33	2:00	3:08
35	2/28/00	Instrument Comparison	Gas	Total	17:25	17:35	18:05	18:49	0:30	1:14
36	2/28/00	Instrument Comparison	Gas	Total	19:20	19:30	20:01	20:58	0:31	1:28
37	2/29/00	Instrument Comparison	Gas	Total	--	18:38	19:41	--	1:03	1:03
38	2/2/00	Oven Dirtying	Gas	Total	20:23	20:33	22:03	23:04	1:30	2:31
38R	2/23/00	Oven Dirtying	Gas	Total	20:06	20:16	21:16	22:18	1:00	2:02
39	2/9/00	Oven Dirtying	Electric	Total	14:14	14:31	15:31	16:35	1:00	2:04

Table 2-3. Summary of the Cooking Protocols

Test Number	Summary Description of Cooking Protocols
1R	Oven Cleaning – Test was performed by running test 38R prior to this test. Oven lock was engaged and the oven was set to the clean setting. The oven was operated for 4 hours.
2, 9, 17, 21	Stovetop Stir Fry – Using a stovetop, 16 g of peanut oil was added and heated for one minute on high. Two batches of pre-packaged frozen vegetables were fried in peanut oil sequentially in two batches and removed. Boneless skinless chicken breasts cut into 2-inch pieces were fried in peanut oil sequentially in two batches, removed, and weighed. In total, 65 g of peanut oil was used.
3, 10, 18,	Bacon – Using the skillet, 6 to 8 pieces of sliced bacon were placed into the pan and cooked turning once. Cooked bacon was removed, and 6 to 8 pieces of bacon was added to pan and cooked in drippings from first batch. Cooking continued until the pound of bacon was cooked. Bacon was removed and served.
4, 11	Tortillas – Using the skillet, 1075 g of vegetable oil was added to the pan and heated for 10 minutes on medium high. Individual white corn flour tortillas were cooked in oil for 1 to 2 minutes each, until golden brown. A total of 24 tortillas were cooked.
5, 12, 22	French Fries – Using cast iron kettle with fryer basket, 3045 g of vegetable oil was added and heated on high until oil temperature reached 190 °C. The contents of one 2-lb bag of steak cut French fires were put into the basket and cooked for 9 minutes. After the first batch of French fries was cooked, a second bag of French fries was cooked.
6, 13, 19	Broil Fish – Using the Pyrex baking dish, one Atlantic salmon steak was placed in the center and brushed with 15 g of extra virgin olive oil. The broiler was heated for 5 minutes, and the steak was placed under the broiler. The steak was cooked for 5 minutes, then removed, turned over, and brushed with 15 g of oil. The steak was cooked for an additional 5 minutes, removed, and served.
7, 14	Bake Lasagna – Using the pan provided by the packaging, one frozen pre-packaged lasagna was removed from the freezer. The oven was turned on to 375 °F and pre-heated for 30 minutes. The lasagna was placed in the center of the oven and cooked for 1.5 hours, removed, and served.
15	Microwave Bacon – Using the Pyrex baking dish and paper towels, 6 strips of sliced bacon was placed on two paper towels in the dish and covered with 2 paper towels. The dish was placed in the microwave and cooked on the highest setting for 15 minutes. The bacon and towels were removed and the test repeated with 6 more strips of bacon.
16	Microwave Lasagna – Using the pan provided by the packaging, one frozen pre-packaged lasagna was removed from the freezer. The lasagna was placed in the center of the microwave and cooked on the highest setting for 30 minutes, turning 90 degrees every 10 minutes.
20	Full Meal – Consisted of baked potatoes, fried chicken, boiled vegetables, boiled broccoli, brown gravy, and rolls. Oven was pre-heated to 400 °F for 30 minutes. Potatoes were poked with knife and placed in Pyrex baking dish. The potatoes were cooked for 1 hour 50 minutes. Yellow squash was prepared by slicing the squash into ½ inch slices and adding one diced yellow onion to the 4-quart pot. Water was added to cover the vegetables. The squash and onions were covered and simmered for 50 minutes. Broccoli was prepared by cutting the florets from the stem, placing them in a 4-quart pot and covering with water. The broccoli was covered and simmered for 50 minutes. The fried chicken was prepared by covering the chicken pieces with a fryer breading mix and cooking in 3390 g of vegetable oil in the cast iron kettle with fryer basket. One half of the chicken was cooked at a time for 20 minutes. The gravy was prepared from a pre-packaged dehydrated brown gravy mix. One cup of water was added to a 4-quart pan and heated for one minute. The gravy mix was added and cooked while stirring for 1 minute. The biscuits were prepared by heating a package of frozen rolls for 23 minutes in the oven during the end of the potatoes cooking time. Food cooking was timed to have all of the food cooking end times within a few minutes of each other.
24, 26, 28, 31	Fry Beef – Using the skillet, ground beef was browned. The skillet was pre-heated for 1 minute; the ground beef was added to the pan and stirred for 1 minute. The ground beef was cooked for 20 minutes, stirring every 5 minutes.
25,27,29	Pork Roast – Using the Pyrex baking dish, the center cut pork roast was placed fat side up in the dish. The oven was pre-heated to 275 °F for 30 minutes, and the roast was placed in the center of the oven. The roast was cooked for 2.5 hours
30	Stovetop Stir Fry – Same as test 2 except used vegetable oil rather than peanut oil.
32	Microwave Popcorn – This test was performed by taking one bag of microwave popcorn and placing it in the center of the microwave oven. The oven was operated on maximum power for 4 minutes
33	Burner Background – Performed by boiling water for 1 hour in a 4-quart pot.
34	Oven Background – Performed by setting oven to bake at 375 deg F for 2 hours.
38R, 39	Oven Dirtying – These tests were performed by spreading a tomato puree and cherry pie filling mixture on the bottom of the oven and cooking oil was spread on the walls, top and bottom of the oven. A total of 90 grams of material was used to dirty the oven prior to oven cleaning. The oven was set to 350 °F for 1 hour to allow the material to bake on.

Note: No spices were used in these preparations.

2.4 Measurement Methods and Performance

2.4.1 Test House and Appliance Measurement Parameters

A laboratory was set up in the garage of the test house. Signal lines were routed from the DAS to temperature and relative humidity probes located in the rooms of the house and outdoors. Signal lines were also routed from the appliances (e.g. for thermocouples in the oven) to the DAS. Tubing for collection of air samples was routed from the garage laboratory to the rooms. Sampling and monitoring inlets were at a standing breathing height of approximately 1.5 m (60 inches). An enclosure was set up in the back yard for temperature and RH sensors.

The Test House was set up for continuous measurements of temperature and RH indoor and outdoors, continuous CO and NO_2 monitoring indoors and outdoors (described in a following section), and automated measurements of air exchange rates by the SF_6 tracer gas decay method. Automated sampling systems were used for collection of air samples from multiple locations. A DAS was used for recording data from the monitors.

The test house and appliance measurement parameters and instrumentation are summarized in Tables 2-4 and 2-5.

Temperature and RH were measured continuously in the kitchen, living room, and master bedroom. Probes consisting of resistance temperature detectors (RTDs) were used for temperature measurements. Thin-film capacitance sensors were used for RH. Temperature and RH were also measured outdoors at the location indicated on Figure 2-1. The outdoor probe was appropriately shielded.

Air exchange rates were measured using an automated SF_6 tracer gas decay method. The method was based on ASTM method E-741 (ASTM, 1999). The tracer gas was introduced into the house manually by releasing 50 mL of pure SF_6 throughout the house, with the volume introduced into each room weighed for the room volume. Dosing was performed approximately 30 minutes before the start of each test. Air was sampled sequentially from the kitchen, living room, master bedroom, and outdoors with an automated system to obtain an average air exchange rate for the house. Analysis was performed with a Hewlett Packard gas chromatograph (GC) with an electron capture detector (ECD). The GC was calibrated with SF_6 standards over a range of 0.5 to 100 ppb. Zero and span checks were performed prior to the start of each day of testing.

Temperatures were measured with thermocouples interfaced to the DAS. Temperatures measured during the tests included the temperature of the flame of the stovetop burner, oven flame temperature and oven compartment temperature, and temperature of cooking oils and/or temperature of foods (e.g., roasts). Placement of the probes in the foods and the flame was difficult to reproduce, making it difficult to compare temperatures between the tests. The volume of gas used in each cooking test was measured with a dry gas meter. The local natural gas supplier reported the BTU content of the gas as 1000 ± 20 Btu/ft^3. During tests with the electric stove, a similar set of temperatures was measured. Electric power was measured with a power transducer (lights and clocks were disabled). Temperature was not measured in the microwave.

Table 2-4. Test House Environmental Measurement Parameters and Instrumentation

Parameter	Instrument	Measurement Principle/Method	Measurement Range	Locations[a]
Temperature	HyCal HT-2W-C-D9-TT-B	Resistance Temperature Detector	-17 to 37.7 °C	K, LR, MBR, OA
Relative Humidity	HyCal HT-2W-C-D9-TT-B	Thin Film Capacitance	0-100 %	K, LR, MBR, OA
Air Exchange	Hewlett-Packard 5890	Tracer Gas (SF_6) Decay w/ GC/ECD	0-100 ppbv	K, LR, MBR, OA
Air Flow Rates	Shortridge ADM 860	Pitot tube	25-10,000 fpm	Air handler
Exhaust Air Flow Rates	Solomat MPM 500	Anemometer (hot wire or vane)	30 - 2500 fpm	Range hood
Data Acquisition				
LabTech Notebook for Win '95	DAS computer	12 bit A/D	Hardware dependent	Inputs from all locations
8-channel A/D board	CyberResearch CYDAS 8	8 channel, 12 bit A/D, 20 kHz	±5 V, 0-10 V	DAS
32-channel multiplexing panel	CyberResearch CYEXP 32	Voltage, current, or thermocouple	switchable gains	DAS
Dosing & Sample Acquisition				
SF_6 Dosing		Manual release of pure SF_6	Not applicable	All rooms with weighted release volumes
SF_6 Sampling System	ARCADIS-configured - poly-line, valves/manifold	Sequential sampling	Not applicable	K, LR, MBR, OA
NO_2/CO Sampling System	ARCADIS-configured - Teflon® line, valves/manifold	Sequential sampling	Not applicable	K, LR, MBR, OA

[a] Locations: Kitchen (K), Living Room (LR), Master Bedroom (MBR), Outdoor Air (OA)

Table 2-5. Appliance Measurement Parameters and Instrumentation

Parameter	Instrument	Measurement Principle/Method	Measurement Range	Locations
Cooking Oil or Food Temperature	K type thermocouple	emf, Seebeck Effect	-200 to 1250 °C	Range top
Flame Temperature	K type thermocouple	emf, Seebeck Effect	-200 to 1250 °C	Range-top burner
Food Temperature	K type thermocouple	EMF, Seebeck Effect	-200 to 1250 °C	Oven
Oven Temperature	K type thermocouple	EMF, Seebeck Effect	-200 to 1250 °C	Oven
Natural Gas Volume	Dry Gas Meter	Positive displacement		Gas range
Electrical Power	Power Transducer	Hall Effect		Electric range

2.4.2 Pollutant Measurement Parameters and Instrumentation

The pollutant sampling and analysis methods are summarized in Table 2-6 and described below. The performance of the measurement methods and instrumentation is summarized in Section 3.10 and highlighted below.

2.4.2.1 Carbon Monoxide (CO) Monitoring

CO was monitored with a Thermo Environmental Instruments Model 48 non-dispersive infrared (NDIR) monitor located in the garage laboratory. The monitor is a bench top sized instrument that is an approved Federal Reference Method (FRM) monitor for National Ambient Air Quality Standards (NAAQS) ambient air monitoring. The monitor was set up to sample from an automated sampling valve and manifold system to sequentially measure CO in air samples from the outdoors (OA), kitchen (K), living room (LR), and master bedroom (MBR). Teflon® sample collection tubes were plumbed from the garage laboratory to each sampling location in the house. To allow for sufficient instrument rise time, the sampling time for each location was 3.75 minutes, which resulted in four measurements at each location per hour. Data from the monitor were recorded with the DAS. Data were processed with a routine that averaged the monitor readings only for the last minute of the measurement cycle. The average during the minute period was saved to the DAS.

Certified gas standards in gas cylinders were used with a dilution system to perform weekly multi-point calibrations over a range from 0 to 20 ppm. Zero and span checks were performed at the start of each day of testing.

The CO monitor, being an FRM, was appropriate for measurements during this study. The detection limit of 0.1 ppm was adequate for the study. The performance of the instruments was excellent based on daily zero and span checks. The bias was less than 2.1 % for all 18 span checks. The relative standard deviation for the span checks performed with a 19.4 ppm standard was 1.0 %.

2.4.2.2 Nitric Oxide (NO) and Nitrogen Dioxide (NO$_2$) Monitoring

NO and NO$_2$ were measured with a Thermo Environmental Instruments Model 42 chemiluminescent monitor located in the garage. The instrument is also a FRM. The monitor was set up in the same manner as the CO monitor, with samples supplied to one instrument with the sampling system that alternated between locations. Teflon® sampling lines were used for transferring the sample to the instrument.   The instrument was calibrated over a range of 0 to 1000 ppb with certified gas standards in gas cylinders and a dilution system. Zero and span checks were performed at the start of each day of testing. Measurements for NO may be truncated to 1000 ppb due to scale limitations during monitoring.

Table 2-6. Pollutant Measurement Parameters and Instrumentation

Parameter	Instrument	Measurement Principle/Method	Measurement Range	Minimum Detection Limits	Locations[a]
Gases - Continuous					
Carbon Monoxide - Area Monitor	Thermo-Environmental Instruments, Inc. Model 48	Non-dispersive Infra-red (NDIR)	1-20 ppm	0.1 ppm	K, LR, MBR, OA (Sequential)
Nitric Oxide (NO_x) - Area Monitor	Thermo-Environmental Instruments, Inc. Model 42	Chemiluminescence Detector	0-1,000 ppb	0.50 ppb	K, LR, MBR, OA (Sequential)
PM (Continuous) - Range from 0.03 μm to 10 μm)	Dekati Electrical Low Pressure Impactor	Multi-stage impactors with multi-channel electrometers	Size range dependent	$0.56/cm^3$ (6.5 μm) $381/cm^3$ (0.047μm)	Kitchen
$PM_{2.5}$ (Integrated) - Personal	Personal Environmental Monitor (PEM)	Impactor/ Gravimetric	Not applicable	17 μg/m^3	Kitchen
$PM_{2.5}$ (Integrated) - Indoor rooms, outdoors	MS&T Sampler	Impactor/ Gravimetric	Not applicable	3 μg/m^3	LR, MBR, OA
PM_{10} (Integrated) - Personal	Personal Environmental Monitor (PEM)	Impactor/ Gravimetric	Not applicable	17 μg/m^3	Kitchen
PM_{10} (Integrated) - Indoor rooms, outdoors	MS&T Area Sampler	Impactor/ Gravimetric		3 μg/m^3	LR, MBR, OA
Polyaromatic Hydrocarbons (PAHs)	Low Flow SKC Pump	Quartz filter/GC/MS[b]	Not applicable	0.4 ng/m^3	K, OA
Elements (PM_{10})	Same as Particles, Integrated	Filter/XRF[c]	Not applicable	Element dependent	K, OA
Aldehydes	Low Flow SKC Pump	DNPH-Silica gel/HPLC[d]	Not applicable	0.5 μg/m^3	K, OA

[a] Locations: Kitchen (K), Living Room (LR), Master Bedroom (MBR), Outdoor Air (OA)
[b] Gas chromatography/mass spectrometry
[c] X-ray fluorescence
[d] High performance liquid chromatography

The chemiluminescent monitor, being a FRM, was appropriate for measurements during this study. The instrument was operated on the 0 – 1000 ppb range. The detection limit of 0.5 ppb was adequate for the study. The performance of the instrument used to measure the concentrations from the four locations was excellent based on daily zero and span checks. The bias measured with the 1000 ppb standard was less than 3% for NO and 8% for NO_2 for all span checks. The relative standard deviation for the span checks was 1.6 % for NO and 4.3% for NO_2.

2.4.2.3 Real-Time Particle Monitoring

Real-time measurements of particle concentrations were performed during the cooking tests using a Dekati Electrical Low Pressure Impactor (ELPI) that is distributed by TSI Particle Instrument Division (St. Paul, MN). The instrument was well suited for the project because it enables real time particle size distribution (particle counts) and concentration measurement ($\mu g/m^3$) in the size range from 30 nm up 10 μm. The instrument has been used in recent studies of PM emissions from commercial kitchen cooking appliances (Gerstler et al., 1998) and indoor sources such as candles and incense (Guo et al., 2000). The ELPI is based on combining electrical detection principle with low-pressure impactor size classification. The electric current carried by charged particles into each of the 12 impactor stages is measured in real-time by a multichannel electrometer. The mid-points of the size fractions measured are 0.04, 0.08, 0.13, 0.21, 0.32, 0.51, 0.81, 1.3, 2.0, 3.2, 5.2, and 8.4 μm.

The ELPI is a bench top size instrument that requires a large vacuum pump for operation. The exhaust of the ELPI vacuum pump was filtered through a high efficiency particulate air (HEPA) filter. The instrument and pump were placed on a cart so that the ELPI could be moved between the kitchen and outdoors. However, the instrument is not very portable. The ELPI was generally located in the kitchen at a location immediately adjacent to the inlets of the integrated PM samplers. The inlet was at the approximate cook breathing height at 3 feet from the front of the stove. To avoid additional air mixing in the kitchen, the ELPI exhaust was ducted in a plastic tube through a gap in the rear sliding door to the outdoors; the gap was filled with a foam strip. The instrument was moved onto the back deck of the house prior to each day's tests and at the end of the tests to perform short term outdoor air measurements. The only location measured indoors was in the kitchen. The instrument was calibrated by the manufacturer. No additional calibrations or performance checks could be performed with the instrument in the field. The instrument was zeroed at the start of each day of testing. The impactor stages were cleaned according to manufacturers instructions.

Data were collected as one-minute averages. Data were collected for at least ten minutes prior to the start of each test to measure the background in the kitchen.

Although there were few operational problems with the instrument, there were short periods during some tests when the electrometers did not appear to operate properly, despite routine cleaning and zeroing of the instrument. Most of the apparent electrometer problems occurred for the larger impactor stages. These problems may have been related to the nature of the aerosol (grease droplets) or due to the elevated moisture levels in the kitchen during cooking of some foods. There were few particles in the three largest size fractions, further reducing the accuracy of the measurements.

The ELPI manufacturer provides an internal algorithm for estimating the mass concentrations for each measurement channel. The mass was estimated using a density of 1.0 g/cm^3. Mass estimates were reasonable for smaller particles based on comparisons to the $PM_{2.5}$ gravimetric data. But the mass estimates for the three channels measuring the larger particles were generally in poor agreement with the PM_{10} gravimetric mass data and were frequently unrealistically high. This may have been due to the low number of large particles and poor counting statistics for these size fractions with a one-minute averaging time.

Guo et al. (2000) reported that mass estimated by the ELPI for the size fractions representing $PM_{2.5}$ could be related to $PM_{2.5}$ gravimetric mass measurements for tests measuring emissions from candles in the EPA Indoor Air Research House. They reported that the correlation could be determined empirically as:

$$\ln C_{grav} = 0.829 \ln C_{ELPI} - 0.475,$$

where C_{grav} is the gravimetric measurement and C_{ELPI} is the estimated $PM_{2.5}$ mass from the ELPI measurement. They reported an R^2 of 0.967 for N = 14.

An attempt was made to determine a relationship between ELPI and gravimetric measurements during the cooking tests in this study, but there was no correlation between the methods. During some tests, the ELPI concentrations were higher than gravimetric. But in other tests, the gravimetric concentrations were higher. The relationship between the gravimetric measurements and the ELPI mass estimates for particles in the $PM_{2.5}$ size fraction are depicted in Figure 2-4.

Figure 2-4. Relationship Between $PM_{2.5}$ Gravimetric Measurements and ELPI Measurements
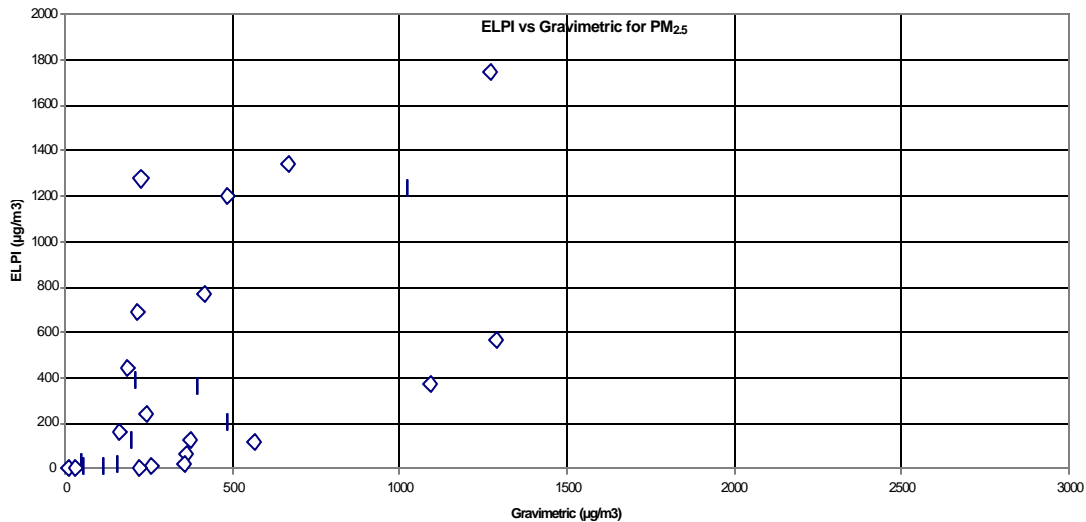
2.4.2.4 Integrated Sampling for $PM_{2.5}$ and PM_{10}

Particle mass in the $PM_{2.5}$ and PM_{10} size fractions was collected with the personal environmental monitors (PEMs) previously used in PTEAM (Thomas et al, 1993) and the MS&T size selective impactor that has been used in the EPA Large Building Studies (EPA, 1994), the EPA Duct Cleaning Study (Fortmann et al, 1997) and other indoor air studies. Both sampling inlets have been described in the literature (Ozkaynak et al., 1996; Turner et al., 2000). The PEM is a small personal sampler that operates at either 4 or 10 L/min. The inlets of the 4 L/min version used in this study have 10 holes that direct the flow toward an oil-coated, sintered metal impactor ring. After impaction, the remaining particles are drawn to the membrane filter in the inlet base. The MS&T Area Sampler draws the air sample through an inlet and nozzle. After the air passes through the nozzle, sized for either $PM_{2.5}$ or PM_{10} cutpoints, the air impacts on an oiled sintered metal impactor plate. For both inlets, the samples were collected on tared Teflo® membrane filters.

$PM_{2.5}$ and PM_{10} were collected with the PEMs in the kitchen. The MS&T samplers were used in the LR, MBR, and outdoors where the concentrations were expected to be lower. The MS&T samplers operated at 20 L/min. Following sampling, filters were stored in a freezer at -10 °C. To minimize volatilization of PM, samples were transported to the lab in coolers using ice packs.

Particle mass was determined gravimetrically. Weighing was performed in a controlled environment weighing facility using a microbalance with a 1 µg resolution. Filters were conditioned in the facility for 48 hours prior to weighing, which was performed according to the protocol developed for the EPA Large Building Study (U.S. EPA, 1993). The method detection limit (MDL) for the ARCADIS weighing protocol with the microbalance in a controlled environment facility is 8 µg mass on the filter, which results in a MDL of 3 $\mu g/m^3$ for a 2-hour sample collected at 20 L/min.

2.4.2.5 PAH Sampling and Analysis Method

PAHs were collected in a subset of tests. Samples were collected outdoors and in the kitchen. The pre-test results indicated that the concentrations in the living room were not substantially different than the kitchen concentrations. The sample collection method involved collection with the PM_{10} size selective inlet containing a quartz filter for collection of particulate matter, followed by an in-line sampler (between the filter and pump) containing XAD-4/PUF. Samplers were operated at the required flow rate for the size cutpoint (4 L/min for the PEM in the kitchen and 20 L/min for the MS&T sampler outdoors). Following sampling, filters and sampling media were stored in a freezer at -10 °C. To minimize volatilization of material, samples were shipped to the lab in ice-packed coolers.

PAH analyses were performed by the Desert Research Institute (DRI) using a Gas Chromatography/Mass Spectrometry (GC/MS) method (Zielinska et al., 1998). Prior to extraction, the following deuterated internal standards were added to each filter-sorbent pair: naphthalene-d_8, acenaphthylene-d_8, phenanthrene-d_{10}, anthracene-d_{10}, chrysene-d_{12}, pyrene-d_{10}, benz[a]anthracene-d_{12}, benzo[a]pyrene-d_{12}, benzo[e]pyrene-d_{12}, benzo[k]fluoranthene-d_{12},

benzo[g,h,i]perylene-d_{12}, coronene - d_{12}, and 1-nitropyrene-d_{11}. The filter-XAD pairs were microwave extracted with dichloromethane. The extracts were then concentrated by rotary evaporation at 20 °C under gentle vacuum to ~1 ml and filtered through 0.45 mm Acrodiscs (Gelman Scientific), rinsing the sample flask twice with 1 ml CH_2Cl_2 each time. Approximately 100 µl of acetonitrile was added to the sample, and CH_2Cl_2 was evaporated under a gentle stream of nitrogen. The final volume was adjusted to 100 µl.

The samples were analyzed by the EI (electron impact) GC/MS technique. A Varian Star 3400CX GC equipped with an 8200CX Automatic Sampler and interfaced to a Varian Saturn 2000 Ion Trap was used for these analyses. Injections (1 µL) were made in the splitless mode onto a 30 m 5% phenylmethylsilicone fused-silica capillary column (DB-5ms, J&W Scientific). Quantification of the individual compounds was obtained by selective ion storage (SIS) technique, monitoring the molecular ion of each compound of interest and the corresponding deuterated internal standard, added prior to extraction.

Calibration curves for the GC/MS quantification were made for the most abundant and characteristic ion peaks of the PAH compounds using the deuterated species most closely matched in volatility and retention characteristics as internal standards. Authentic PAH standards (purchased from Aldrich, Inc.) plus National Institute of Standards and Technology (NIST) Standard Reference Material (SRM) 1647 (certified PAH) with the addition of deuterated internal standards were used to make calibration solutions.

A three-level calibration was performed for each compound of interest, and the calibration check (using median calibration standards) was run every ten samples to check for accuracy of analyses. If the relative accuracy of measurement (defined as a percentage difference from the standard value) was less than 20%, the instrument was recalibrated.

MDLs for the PAHs were estimated to range from 3 ng/m^3 for naphthalene to 6 ng/m^3 for coronene assuming a 1 L sample volume. Sample volumes in this study ranged from 0.75 to 1.9 m^3. The DRI laboratory does not include the MDL for each PAH in each sample, but calculates an uncertainty level for each compound in the sample. The uncertainty is calculated as follows:

1. Determine the replicate precision by comparing the same compound result (ng/sample) in replicate injections. The replicate precision is calculated as a fraction (difference/mean of the two values) and averaged over all replicates.
2. Then compute the root-mean-square of (A) the replicate precision from step 1 multiplied by the value for which the uncertainty is being determined, and (B) the analytical detection limit.

The effect of this calculation is that the uncertainty is equal to the MDL if the measured value is zero, and the uncertainty becomes increasingly small as the sample concentration gets very high. In this report, all concentrations are reported, but the values below the uncertainty level calculated by DRI are italicized.

2.4.2.6 Elemental Analyses

Elemental analyses were performed for particles collected in the PM_{10} fraction during a subset of tests. Samples were analyzed by Desert Research Institute with XRF using a Kevex Corporation Model 700/8000 energy dispersive x-ray fluorescence (EDXRF) analyzer. Analyses were performed following the DRI Standard Procedure No. 2-205.2. The method is the same as used in the previous ARB study of air pollutants inside California vehicles (Rodes et al., 1998). The estimated detection limits for a 2.4 m^3 volume sample (20 L/min for 120 minutes) are listed in Table 2-7. The MDL for samples collected with the PEMs would be five times higher. As was the case for PAHs, DRI no longer reports the MDL, but they report the uncertainty associated with each measurement instead. Therefore, all element concentrations are presented in this report, with concentrations below the uncertainty level presented in italics.

2.4.2.7 Aldehydes

Samples were collected during a limited number of tests for determination of selected aldehydes. Air samples were collected on silica gel cartridges coated with acidified 2,4-dinitrophenylhydrazine (DNPH). The method is described in the *EPA Compendium of Methods for the Determination of Air Pollutants in Indoor Air* (U.S. EPA, 1999). The commercially available cartridges (Waters Sep-Pak DNPH Silica Gel Cartridge, Waters Associates, Milford, PA) contain 2.9 grams of a 55 to 105 μm chromatographic-grade silica gel. Samples of 40 to 140 L volume were collected with a vacuum pump and mass flow controller at nominal sampling rates of 0.2 to 0.3L/min. The sampling volumes were well below the maximum volumes recommended in the EPA Compendium Method TO-11A (U.S. EPA, 1999). The sampling flow rates were measured at the start and end of each collection period.

Samples collected on DNPH-coated silica gel cartridge were extracted with 5 mL of acetonitrile (UV grade). Twenty-five μL of the extract was then analyzed with a HP 1090 HPLC equipped with a diode array detector. Chromatography was performed with a C-18 reverse phase column (Zorbax ODS, 4.6 x 250 mm) using a gradient program [0 - 10 min at 60 % acetonitrile (ACN) and 40% water, 10 - 20 min at 70% ACN and 30% water, 20-25 min at 100% ACN, and 25-41 min at 60 % ACN and 40% water].

Table 2-7. Minimum Detection Limits for Elements Analyzed by XRF

Element	ng/m³[a]	Element	ng/m³[a]
Al	44.8	Br	4.5
Si	28.2	Rb	4.5
P	25.1	Sr	4.9
S	22.4	Y	5.8
Cl	44.8	Zr	7.6
K	27.3	Mo	12.1
Ca	20.2	Pd	49.3
Ti	13.0	Ag	53.8
V	11.2	Cd	53.8
Cr	8.5	In	58.2
Mn	7.2	Sn	76.1
Fe	6.7	Sb	80.6
Co	3.9	Ba	232.9
Ni	4.0	La	277.7
Cu	4.9	Au	13.9
Zn	4.9	Hg	11.6
Ga	8.5	Tl	11.2
As	7.2	Pb	13.4
Se	5.4	U	10.3

[a] Based on a 2.4 m³ sample volume on a 37 mm filter

The HPLC was calibrated for seven carbonyl compounds: formaldehyde, acetaldehyde, propanal, butanal, benzaldehyde, pentanal, and hexanal. The target compounds were identified by comparison of their chromatographic retention times with those of the derivatized standards. Quantification was performed using an external standard method with a five-point calibration based on peak area of derivatized standards. Standards were prepared at five concentration levels (between 0.04 and 1.50 ng/µL), and a calibration curve was generated by linear regression treatment of the concentration and chromatographic response data. Performance of the instrument was verified on each day of analysis by analysis of a calibration check sample prior to analysis of samples. The practical quantitation limit, which was based on the lowest calibration standard was 3.9 µg/m³ for a nominal 45 L sample volume. The MDL was 0.5 µg/m³ for a 40 L sample. None of the target compounds were detected in the field blanks.

2.5   Data Analysis and Emission Rate Calculation

2.5.1   Analysis Objectives and Analysis Plan

The major analysis objectives for the study are summarized in Table 2-8. Although there were 32 cooking tests performed in the main study, there was a minimal amount of replicate testing. The design of the study was such that it addressed many different variables to characterize the range of the resultant emission rates and exposures.   Due to the limited number of replicates, the significance of differences between the tests is difficult to determine. It should be recognized

that the analysis of the data consists almost exclusively of preparation of the summary statistics and comparison of summary data from different tests or groups of tests. The data set was too small to perform more complicated analyses. Much of the discussion in Section 4.0 is descriptive.

2.5.2 Summary Statistics

The summary statistics presented in this report are the basic statistical techniques and include the calculation of averages, standard deviations, medians, minimums, and maximums.

For real-time data on CO and NO_2, the data were analyzed to obtain peak and average concentrations during the cooking period and the total exposure period (duration of cooking plus time period after termination of cooking, as specified in the cooking protocol).

Table 2-8. Data Analysis Objectives and Plan

Program Objective	Analysis Plan
Measure personal exposures and indoor concentrations for $PM_{2.5}$ and PM_{10} during cooking	Calculate average and variance of concentrations during cooking exposure period at all locations in the house
Measure personal exposures and indoor concentrations for CO and NO_2 during cooking	Compute peak, average, and standard deviation of concentrations of pollutants at all locations
Measure personal exposures and indoor concentrations for PM in size fractions from 0.1 to 10 μm	
Measure personal exposure and indoor concentrations of PAHs, elements, and aldehydes during cooking	Compute peak, average, standard deviation, and median concentrations during cooking and exposure periods
Compare personal exposures and indoor concentrations under typical and worst-case conditions during cooking	Determine differences between indoor concentrations and emission rates
Determine emission rates of PM, CO, and NO_2 produced by cooking under typical conditions	Calculate emission rates using mass balance model
Determine emission rates of PM, CO, and NO_2 produced by cooking under worst-case conditions	Calculate emission rates using mass balance model
Evaluate impact of appliance type (gas range, electric range, microwave) on pollutant emission rates	Determine differences in indoor concentrations and emission rates
Evaluate impact of the type of cooking method on pollutant emission rates	Determine differences in indoor concentrations and emission rates
Evaluate impact of food type on pollutant emission rates	Determine differences in indoor concentrations and emission rates
Evaluate impact of cooking utensil material (iron or aluminum) on pollutant emission rates	Determine differences in indoor concentrations and emission rates
Evaluate impact of exposure reduction methods on cook exposure and indoor concentrations	Determine differences in indoor concentrations and emission rates

2.5.3 Emission Rates

Although the primary objective of the study was to measure exposures to air contaminants due to cooking in the test house, the technical approach was developed such that emission rates and source strengths could be calculated using a dynamic mass balance model. Emission rates were calculated for $PM_{2.5}$ and PM_{10}, measured by the gravimetric method, CO, NO, and NO_2, and for selected size fractions measured with the ELPI. A dynamic mass balance model was used, as follows:

$$ER = \dfrac{V(a+k)T_e\left(C_i - \dfrac{fa}{a+k}C_o\right)}{T_c - \dfrac{1}{(k+a)}\left[(e^{(-(k+a)(T_e-T_c))}) - (e^{(-(k+a)T_e)})\right]}$$

Where:

ER = emission rate (µg/hr)

V = house volume (m^3)

a = air exchange rate (hr^{-1})

k = pollutant deposition or decay rate (hr^{-1})

C_i = indoor kitchen concentration ($µg/m^3$)

f = penetration factor (unitless)

C_0 = outdoor concentration ($µg/m^3$)

T_e = total exposure time (hr)

T_c = total cooking time (hr)

Parameters in the model that were measured included C_i, C_0, air exchange rate, and house volume. The value f was set to one, assuming complete mixing in the house. The penetration factor was set to 1 for all parameters. The deposition rates were set equal to 0.0 hr^{-1} for CO and NO. A deposition rate of 0.8 hr^{-1} was used for NO_2, based on data collected previously for ARB model development (Koontz et al., 1998) and published data compiled by Traynor (1999) for the California Department of Health Services Indoor Air Quality Section.

The deposition velocity for $PM_{2.5}$ was set to 0.31 hr^{-1}. This rate was determined based on the differences in the decay rates of $PM_{2.5}$ mass concentrations measured with the ELPI data and SF_6 decay rates measured to calculate air exchange rates in 14 representative tests that covered a range of cooking activities. The rate was in reasonable agreement with the rate of 0.39 hr^{-1} reported by Wallace (1996) from PTEAM. The rate of 1.01 hr^{-1} reported by Wallace from PTEAM was used for the PM_{10} and elemental deposition rate because the ELPI data for the larger size fractions were inadequate to estimate the deposition rate during the cooking tests in this study. A deposition velocity of 1.01 hr^{-1} was also used for the elements analyzed in samples of PM_{10}.

Data on deposition velocity for PAHs, aldehydes, and elements were not available. A deposition velocity of 0.0 was used for calculating the emission rates of PAHs, consistent with that used by Sheldon et al. (1992) in a previous ARB project. Due to the lack of data, a deposition velocity of 0.0 was also used for aldehydes.

Source strengths were calculated as µg of pollutant emitted per g of food cooked and per unit of energy (µg/BTU). Emission rates were also calculated as µg of pollutant emitted per g of food per hour (µg/g/h).

Some mass balance model calculations resulted in negative values because the outdoor concentration was higher than in the kitchen. The primary cause of this was environmental effects. For example - outdoor samples were integrated over a full day's operation while indoor samples were taken only during each cooking test. The outdoor sample tended to be more accurate because the longer sampling period provided a larger sample, while the shorter period indoor samples yielded smaller catches or values closer to the method detection or quantification limits. In addition, for some constituents, there was no measurable difference between outdoor and indoor samples because the indoor cooking emissions were balanced by the effects of multiple traffic rush hours, woodstove heating cycles, surrounding residential cooking, etc. on the day-long, average ambient pollutant levels. A related experimental error occurred when very low measured concentrations were reduced to zero or negative values when corrected for media blank data. Because it is a possibility that outdoor concentrations would exceed indoor concentrations of these pollutants, all resulting negative values are entered into the data tables as flagged zeros.

Peak concentrations for the gaseous emissions were computed using the peak kitchen concentrations and peak outdoor concentrations. This may have resulted in an underestimation of the peak concentrations due to a peak outdoor concentrations possibly not occurring at the same time as the indoor peak concentration. This is seen in only a few cases for CO and NO. Also, some peak measurements of NO were truncated at 1000 ppb due to scale limitations of the monitor.